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54 Additive concentrate for polyester compositions and method of addition.

57 Thermoplastic compositions of a polyester and/or polycarbonate resin, an acrylic core-shell impact modifier and a stabilizer package are provided by an improved method in which the input modifier and stabilizer components are first preblended and the preblend is then admixed with the polyester and/or polycarbonate. The resultant blend composition retains good properties even without the use of high amounts of the stabilizers.

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UNITED STATES PATENT APPLICATION

OF: Surendra Hukamchand Agarwal

FOR: ADDITIVE CONCENTRATE FOR POLYESTER
COMPOSITIONS AND METHOD OF ADDITION

BACKGROUND OF THE INVENTION

High molecular weight linear polyesters and copolyesters of glycols and terephthalic or isophthalic acid have been available for a number of years. These are described, inter alia, in Whinfield, et al., U.S. 2,465,319 and in Pengilly, U.S. 3,047,539, incorporated herein by reference. These patents disclose that the polyesters are particularly advantageous as film and fiber formers.

With the development of molecular weight control, the use of nucleating agents and two-step molding cycles, poly(ethylene terephthalate) has become an important constituent of injection moldable compositions. 5 Poly(1,4-butylene terephthalate), because of its very rapid crystallization from the melt, is uniquely useful as a component in molding resins. Work-pieces molded from such polyester resins, alone or combined with reinforcements, in comparison with other thermoplastics, 10 offer a high degree of surface hardness and abrasion resistance, high gloss, and lower surface friction.

Stable polyblends of poly(1,4-butylene terephthalate) and poly(ethylene terephthalate) can be 15 molded into useful unreinforced and reinforced articles. See Fox and Wambach, U.S. 3,953,394, incorporated herein by reference.

Block copolyesters containing units derived 20 from poly(1,4-butylene terephthalate) and from an aromatic aliphatic or aliphatic polyester are also known. See copending application U.S. Serial No. 290,879, filed August 7, 1981, which is itself a continuation of U.S. Serial No. 752,325, filed December 20, 1976, incorporated 25 herein by reference. Such block copolyesters are useful per se as molding resins and also in intimate combination with poly(1,4-butylene terephthalate) and/or poly(ethylene terephthalate).

30 It has been proposed to increase the impact strengths of polyesters by adding various modifiers. For example, Brinkmann, et al. in U.S. 3,591,659 disclose that a useful family of modifiers comprises polyalkyl acrylates, methacrylates and/or ethacrylates. Baron, 35 et al. in U.S. 4,044,073 disclose that a useful impact

modifier for such polyesters is an aromatic polycarbonate. Schlichting, et al., U.S. 4,022,748, disclose that a rubber-elastic graft copolymer having a glass temperature below -20°C. is a useful modifier. Lane, U.S. 4,034,013, and Farnham, et al., U.S. 4,096,202, disclose that useful impact modifiers comprise multiple stage polymers having a rubbery first stage and a hard final stage, preferably including units derived from alkyl acrylates, especially butyl acrylates. Baron, et al. in U.S. 4,034,016 (corres. German 2,650,870) disclose an impact modifier combination comprising a blend of a polyurethane and an aromatic polycarbonate. Copending application Serial No. 870,679, filed January 19, 1978, discloses an impact modifier combination comprising a segmented block copolymer and an aromatic polycarbonate. Cohen and Dieck, U.S. 4,257,937, disclose an impact modifier combination comprising a blend of a polyalkylacrylate and an aromatic polycarbonate. Gergen, et al., U.S. 4,090,996, disclose an impact modifier combination for poly(1,4-butylene terephthalate) and other polyester resins comprising a selectively hydrogenated monoalkenyl arene-diene block copolymer, and an engineering thermoplastic, e.g., poly(aryl ether), poly(aryl sulfone), polycarbonate, acetal, etc. Nakamura, et al., U.S. 3,864,428, disclose poly(butylene terephthalate) alone or combined in blends with other polyesters, impact modified with conjugated diene/vinyl aromatic copolymers grafted with methacrylate monomer and aromatic polycarbonate. Fromuth, et al., U.S. 4,180,494, disclose poly(butylene terephthalate), a core shell polymer having a butadiene core and up to 8 percent by weight of an aromatic polycarbonate. Dieck, U.S. 4,280,948, discloses poly(butylene terephthalate) copolyesters impact modified with a combination of an acrylic or methacrylic monomer grafted polymer of a conjugated diene alone or combined with vinyl aromatic

and an aromatic polycarbonate resin. All of the foregoing patents and the application are incorporated herein by reference.

5 It is also known that acrylic rubbers, methyl methacrylate-butadiene-styrene terpolymers and, in general, finely divided particulate rubbery materials made by emulsion polymerization are more readily dispersed in a polymer matrix such as poly(1,4-butylene terephthalate)
10 if pre-blended, i.e., concentrated, with a polyolefin or co-polyolefin.

More recently, it has been discovered that the addition of an acrylic or methacrylic monomer grafted
15 core shell polymer of more than 50 weight percent of a conjugated diene, alone or in combination with a vinyl monomer, e.g., Rohm & Haas' Acryloid KM 653, and an aromatic polycarbonate, e.g., General Electric Company's LEXAN[®], to poly(1,4-butylene terephthalate), e.g., General Electric Company's VALOX[®], results in a composition
20 which, when molded into parts, shows good impact strength. While methacrylate-butadiene-styrene copolymers, not of the core-shell type, available from other sources, also give good impact strength at room temperature, e.g.,
25 23°C., only the compositions made from the core shell polymers retain more than 70% of their impact strength at -40°C., making them uniquely suitable for automobile bumpers, ski bindings, and the like, where low-temperature impact resistance is all important. The compositions
30 are described and claimed in application Serial No. 316,946, filed October 30, 1981, having the same assignee as herein. As disclosed therein, in preferred embodiments the high temperature performance, and to an extent the low temperature performance, of the compositions is improved by the
35 addition of antioxidants and stabilizers.

SUMMARY OF THE INVENTION

5 The discovery has now been made that if antioxidants/stabilizers are precompounded with the acrylic core shell impact modifying polymer to form a concentrate, the concentrate provides better dispersion of the core shell polymer and the antioxidants/stabilizers in the final blend composition.

10 For ease of discussion, the antioxidants and stabilizers of the aforementioned copending application will be referred to throughout the present disclosure simply as "stabilizers".

15 This invention essentially involves the pre-blending of the stabilizers and the rubbery core shell polymer under relatively mild conditions of low temperature and low shear. When the resulting concentrate is
20 intimately admixed with the polyester resin, and any other ingredients, at the high processing temperatures normally required to melt the polyester, the stabilizers are already dispersed in the rubbery core shell polymer in relatively high concentration. This results in
25 optimum stabilization of the most temperature sensitive component of the blend composition -- the core shell polymer -- and concomitantly, significantly improved impact strength in the blends after molding, as will be shown in the accompanying examples. Moreover, the
30 concentrate is easier to handle, eliminates or at least sharply reduces the danger of chemical dust explosions and worker exposure to chemical dust.

35 The further discovery has been made that the inclusion of a small amount of a homopolymer or copolymer

of ethylene before or during admixture of the core shell polymer with stabilizers considerably eases preparation of the concentrate.

5

DESCRIPTION OF THE INVENTION

10 In greater detail, in one aspect the invention comprises an improvement in the method of preparing a composition, comprising admixing

(a) a thermoplastic polymer selected from the group consisting of: (1) a polyester which is (i) a poly(1,4-butylene terephthalate) resin, or (ii) a block
15 copolyester of poly(1,4-butylene terephthalate) and an aromatic/aliphatic or aliphatic polyester, or (iii) a blend of (i) and a poly(ethylene terephthalate) resin, or (iv) a blend of (ii) and a poly(ethylene terephthalate) resin; (2) an aromatic polycarbonate resin, and (3) a
20 mixture of (1) and (2);

(b) the balance to make 100 parts of (a) and (b) together of an impact modifier thereof comprising an acrylic or methacrylic monomer grafted core shell polymer of a conjugated diene, alone or in combination with a
25 vinyl monomer, the core being polymerized from over 50 weight percent of said diene; and

(c) a stabilizer composition comprising (i) a hindered phenolic antioxidant, (ii) optionally, an aromatic amine, (iii) a thioester, and optionally, (iv)
30 a trivalent phosphorus compound.

The improvement comprises forming a pre-blended concentrate of impact modifier component (b), i.e., the rubbery core shell polymer, and component (c),
35 the stabilizer composition, then admixing it with the

other described ingredients of the composition.

In still further detail, the preblend concentrate is prepared by dry blending the components (b) and (c) and extruding or otherwise compounding the components at a relatively low temperature of 200 to 450°F., then collecting the compound for addition to the rest of the composition as described above.

In preparing the concentrate, in addition to use of the low temperatures as described, the components are mixed under conditions regulated to provide relatively low shear. The aim is to avoid exposing the core shell resin component to high temperature and/or high shear rates, to prevent the degradation of the rubbery core. The residence time during compounding should also be kept minimum. Prolonged hot roll milling and intensive high shear Banbury mixing are not at all advantageous; short term milling followed by extrusion gives the best results.

In another aspect of the invention, there is provided a preblend concentrate formed in the above-mentioned manner, comprising (1) an intimate admixture of acrylic or methacrylic monomer grafted core shell polymer of a conjugated diene, alone or in combination with a vinyl monomer, the core being polymerized from over 50 weight percent of said diene, and (2) a stabilizer composition of a hindered phenolic antioxidant, optionally, an aromatic amine, a thioester, and, optionally, a tri-valent phosphorus compound.

The polyester resins of (a) of this invention are available commercially or can be prepared by known techniques such as transesterification of esters

of terephthalic acid with ethylene glycol or butanediol and subsequent polymerization, by heating the glycols with the free acids or with halide derivatives thereof, and similar processes. These are described in U.S.

Patents 2,465,319 and 3,047,539, and elsewhere. A suitable poly(1,4-butylene terephthalate) as defined under (a) (1) (i) above is available commercially from General Electric Company, Pittsfield, Massachusetts, under the trade designation VALOX® 303.

The block copolyester resin component used in (a) (1) (ii) above can be prepared by the transesterification of (a) straight or branched chain poly(1,4-butylene terephthalate) and (b) a copolyester of a linear aliphatic dicarboxylic acid and, optionally, an aromatic dibasic acid such as terephthalic or isophthalic acid with one or more straight or branched chain dihydric aliphatic glycols. For example, a poly(1,4-butylene terephthalate) can be mixed with a polyester of adipic acid and ethylene glycol, and heated at 235°C. to melt the ingredients, then heated further under a vacuum until the formation of the copolyester is complete. As the second component, there can be substituted poly(neopentyl adipate), poly(1,6-hexylene azelate-co-isophthalate), poly(1,6-hexylene adipate-co-isophthalate) and the like. The procedures and products are described in the above-mentioned copending application U.S. Serial No. 290,879, filed August 7, 1981, as a continuation of U.S. Serial No. 752,325, filed December 20, 1976.

The poly(ethylene terephthalate) resin component used as part of (a) (1) (iii) and (a) (i) (iv) of the

compositions of this invention is available commercially or can be prepared by known techniques such as by the transesterification of esters of terephthalic acid with ethylene glycol with the free acids or with halide derivatives, and similar processes. These are described in U.S. 2,465,319 and U.S. 3,047,539, and elsewhere.

Illustratively, the high molecular weight polyesters will have an intrinsic viscosity of at least about 0.2 deciliters/gram and, preferably, at least about 0.4 deciliters/gram as measured in a 60:40 phenol/tetrachloroethane mixture at 30°C., for poly(ethylene terephthalate) and at least 0.6 and more, preferably 0.8 deciliters/gram, same basis, for poly(1,4-butylene terephthalate). Most preferably, for the former, the intrinsic viscosity will be in the range of 0.5 to 1.0 dl./g. and from 0.9 to 1.2 for the latter.

Especially useful when high melt strength is important are branched high melt viscosity poly(1,4-butylene terephthalate) resins, which include a small amount, e.g., up to 5 mole percent based on the terephthalate units, of a branching component containing at least three ester forming groups. The branching component can be one which provides branching in the acid unit portion of the polyester, or in the glycol unit portion, or it can be hybrid. Illustrative of such branching components are tri- or tetracarboxylic acids, such as trimesic acid, pyromellitic acid, and lower alkyl esters thereof, and the like, or preferably, polyols, and especially preferably, tetrols, such as pentaerythritol, triols, such as trimethylolpropane; or dihydroxy carboxylic acids and hydroxydicarboxylic acids and derivatives, such as tartaric acid, and the like.

The branched poly(1,4-butylene terephthalate) resins and their preparation are described in Borman, U.S. 3,953,404, incorporated herein by reference.

5 Impact modifier (b) comprises an acrylic or methacrylic monomer grafted polymer of a conjugated diene alone or copolymerized with a vinyl aromatic compound. The graft copolymer resin (b) must be made in such a way to distinguish it from those available
10 commercially from Borg-Warner Corp., under the trade designation BLENDX, and those available from Kanegafuchi Chemical Industry Co. of Japan, under the trade designation of KANE ACE. Good low temperature impact properties are not obtained even with Rohm & Haas material trade
15 designated Acryloid KM 330. In general, only the core shell polymers of the type available from Rohm & Haas under the trade designation Acryloid KM 653 can be used, especially those containing units derived from butadiene or isoprene, alone or in combination with a vinyl aromatic
20 compound. These are believed to be described in the above-mentioned Fromuth, et al. patent. Especially preferably, the copolymer component (b) will comprise a three-stage polymer having a butadiene-based rubbery core, a second stage polymerized from styrene, and a final stage, or shell,
25 polymerized from methyl methacrylate, and 1,3-butylene glycol dimethacrylate.

The polycarbonate resins (2) can be made in known ways and they are available commercially from sources,
30 e.g., General Electric Company, Pittsfield, Massachusetts, U.S.A., under the trademark LEXAN®. In general, any of the aromatic polycarbonates described in Baron, et al., U.S. 4,034,016 can be used, especially those including units deriv d from bisphenol-A.

In preferred cases, the compositions of this invention will comprise from about 30 to about 90 parts by weight of component (a) per 100 parts of total weight of the composition; and from about 5 to about 30 parts by weight of component (b) per 100 parts of the composition.

The preferred stabilizer combination (c) comprises (i) a hindered phenolic antioxidant, (ii) optionally, an aromatic amine, (iii) a thioester, and, optionally, (iv) a trivalent phosphorus compound, e.g., a phosphite in amounts from 0.25 to 1.5 parts each of (i), (ii) and (iii) and from 0.5 to 5 parts of (iv) per 100 parts by weight of (a) and (b) together.

Optionally, the compositions can also include (d) fillers and/or reinforcing agents. Special mention is made of fibrous (filamentous) glass, which when present is generally included in amounts of up to about 60% and more, usually up to about 40% by weight of (a) and (b) combined. However, any filler, e.g., mineral filler such as talc, mica, clay, or fibrous reinforcement, can be used in these amounts.

Other ingredients, such as dyes, pigments, drip retardants, flame retardants, and the like can be added in conventional amounts for their conventionally employed purposes.

As explained previously, the concentrate is admixed with the other ingredients of the final composition. This can be done in one convenient manner by tumbling the concentrate and other components to form a dry blend, extruding the blend at a temperature between 450 to 550°F., cooling the extrudate, forming

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it into pellets and injection molding at a temperature of 450 to 550°F. (50 to 250°F. mold temperature).

5 The molded compositions are useful for a variety of purposes where performance at below normal and/or above normal temperatures is desired, for example, automobile bumpers, ski bindings, and the like.

10

DESCRIPTION OF THE SPECIFIC EMBODIMENTS

The following examples illustrate the invention. They are set forth as a further designation only and are
15 not to be construed as a limitation on the scope of the invention.

EXAMPLES 1-4

20 Dry blends of poly(1,4-butylene terephthalate) resin, aromatic polycarbonate of bisphenol-A and a concentrate of polybutadiene-styrene-methyl methacrylate core shell polymer and stabilizers were compounded and extruded at 266°C. melt temperature in an extruder. For
25 comparison purposes, all of the ingredients were admixed together directly, without prior formation of the concentrate. The extrudates were pelletized and injection molded at 238°C. (mold temperature 66°C.). The formulations and physical properties are shown in Table 1.

30

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TABLE 1

Example	Composition (parts by weight)	Not Pre-Concentrated				Pre-Concentrated			
		1	2	3	4	1	2	3	4
	Poly(1,4-butylene terephthalate) ^a	65	67.45	65	67				
	Aromatic polycarbonate ^b	15	15	15	15				
	S/B/MMA core shell polymer ^c	15	15	--	--				
	Concentrate (1)	--	--	19.5	--				
	Concentrate (2)	--	--	--	16.5				
	Thioester ^d	2.0	0.7	2.0	.19				
	Hindered phenolic antioxidant ^e	0.5	0.35	0.5	.10				
	Aromatic amine stabilizer ^f	0.5	--	0.5	--				
	Aromatic phosphite ^g	0.5	--	0.5	--				
	Polyethylene processing aid ^h	1.5	1.5	1.5	1.5				
<u>Properties</u>									
Notched Izod impact strength,									
ft.lbs./in.									
at 25°C. (room temp.)									
at -30°C.									
		17.0	18.2	16.2	18.2				
		16.6	7.0	17.0	18.2				

1) 100 parts KM-653, 13.3 parts Seenox 412S, 3.3 parts Topanol CA and 3.3 parts Naugard 445 and 10 parts Microthene (FN 510).

2) 100 parts KM-653, 1.14 parts Seenox 412S, 0.6 parts Topanol CA.

a VALOX ® 315, General Electric Company.

b LEXAN ® 134, General Electric Company.

c Acryloid KM-653, Rohm & Haas Chemical Company.

d Seenox 412S, Argus

e Topanol CA, ICI Americas.

f Naugard 445, Uniroyal

g Naugard PHR, Uniroyal

h Microthene (FN 510).

A comparison of test data for control Examples 1 and 2 with the data for pre-blend concentrate Examples 3 and 4 shows that pre-concentration enables one to incorporate into the compositions markedly less stabilizer without adversely affecting low temperature impact strength. These Examples also establish that, without pre-concentration, reducing the stabilizer content has an adverse effect upon low temperature impact strength.

10

EXAMPLES 5-13

Again in accordance with the invention, dry blends of poly(1,4-butylene terephthalate), aromatic polycarbonate, filler/reinforcement and concentrate of S/B/MMA and stabilizers were prepared and extruded through a single screw extruder at 260-288°C. Parts were molded and tested. The formulations and test results are given in Table 2.

20

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TABLE 2

EXAMPLE	5	6	7	8	9	10	11	12	13
<u>Composition (parts by weight)</u>									
Poly(1,4-butylene terephthalate) ^a	65	60	30	60	55	60	55	45	--
Aromatic polycarbonate ⁱ	15	15	15	15	15	15	15	15	90
Concentrate(1)*	18	18	18	18	18	18	18	18	10
Polyethylen ^h	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	--
Aromatic phosphite ^g	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Glass filaments	--	5	--	--	--	--	--	--	--
Clay	--	--	--	5	10	--	--	--	--
Mica	--	--	--	--	--	5	10	--	--
Poly(ethylene terephthalate)	--	--	35	--	--	--	--	20	--
<u>Properties</u>									
Notched Izod Impact Strength, ft.lbs./in., R.T.	15.8	4.5	13.1	7.7	3.4	4.8	3.0	14.1	15
Flexural strength, psi x 10 ⁻³	9.8	10.5	9.8	9.8	9.8	9.6	9.7	9.6	13.2
Flexural modulus, psi x 10 ⁻³	262	296	254	274	303	303	366	255	312
Tensile strength, psi x 10 ⁻³	6.0	6.4	6.2	6.1	6.0	5.7	5.7	5.9	8.2
Elongation, %	161	81	212	143	125	93	10	160	80

* See Table 1, 100 parts KM 653, 13.3 parts Seenox 412S, 3.3 parts Topenol CA, 3.3 parts Naugard 445.

^a As in Table 1.

i) LEXAN 131, General Electric Company

h) As in Table 1.

g) As in Table 1.

It is seen that reinforced materials are obtained from which are produced workpieces with very satisfactory mechanical properties.

5 Obviously, other modifications and variations of the present invention are possible in light of the above disclosure. For instance, the stabilizer package can include an amount of a trivalent phosphorus stabilizer. A copolyester of poly(1,4-butylene terephthalate) and a
10 hydroxy-terminated polyester
 can be substituted for the linear polyester and either can be partially replaced with poly(ethylene terephthalate). All such obvious modifications which
15 may be made in the particular embodiments described above are within the full scope of the invention as defined in the appended claims.

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CLAIMS

- 5 1. An improvement in the method of preparing a thermoplastic composition comprising admixing
- (a) a thermoplastic polymer selected from the group consisting of
- (1) a polyester resin which is
- 10 (i) a poly(1,4-butylene terephthalate) resin, or
- (ii) a block copolyester of poly(1,4-butylene terephthalate) and an aromatic/aliphatic or aliphatic polyester, or
- 15 (iii) a blend of (i) and a poly(ethylene terephthalate) resin, or
- (iv) a blend of (ii) and a poly(ethylene terephthalate) resin;
- (2) an aromatic polycarbonate resin;
- 20 and
- (3) mixtures of (1) and (2);
- (b) the balance to make 100 parts of (a) and (b) together of an acrylic or methacrylic polymer of a conjugated diene, alone or in combination with a vinyl
- 25 monomer, the core being polymerized from over 50 weight percent of said diene; and
- (c) a stabilizer composition comprising
- (i) a hindered phenolic antioxidant,
- (ii) optionally, an aromatic amine,
- 30 (iii) a thioester, and, optionally,
- (iv) a trivalent phosphorus compound,
- said improvement comprising forming a pre-blend of components (b) and (c) and intimately admixing the pre-blend with component (a).
- 35

2. A method according to Claim 1, in which the pre-blend of (b) and (c) is prepared by admixing the two under relatively mild conditions of temperature.

5 3. A method according to Claim 2, in which the temperature is between 93 and 232°C.

4. A method according to Claim 1, in which the pre-blend of (b) and (c) is co-extruded with (a).

10

5. A thermoplastic composition made by the method of Claim 1.

6. A composition as defined in Claim 5,
15 wherein the polyester composition (a) comprises from about 30 to about 90 parts by weight, and impact modifier (b) comprises from about 5 to about 30 parts by weight.

7. A composition as defined in Claim 5, in
20 which stabilizer component (c) is present in amounts of from 0.25 to 1.5 parts by weight of each of (i), (ii) and (iii) and from 0.5 to 5 parts of (iv) per 100 parts of (a) and (b) combined.

8. A composition as defined in Claim 5,
25 wherein said poly(1,4-butylene terephthalate) in component (a) has an intrinsic viscosity of at least about 0.9 deciliters per gram when measured in solution in a 60:40 mixture of phenol and tetrachloroethane at 30°C.

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9. A composition as defined in Claim 8,
wherein said poly(ethylene terephthalate) in component
(a) has an intrinsic viscosity of at least about 0.5
deciliters per gram when measured in solution in a
5 60:40 mixture of phenol and tetrachloroethane at 30°C.

10. A composition as defined in Claim 5,
wherein in said component (a) (1) (i) said poly(1,4-butylene
terephthalate) resin is linear or branched.
10

11. A composition as defined in Claim 10,
wherein said branched polyester is a high melt viscosity
(1,4-butylene terephthalate) resin which includes a small
amount of a branching component containing at least three
15 ester forming groups.

12. A composition as defined in Claim 5,
wherein in graft core shell polymer component (b) (i),
the conjugated diene polymer comprises butadiene or iso-
20 prene, alone or in combination with a vinyl aromatic com-
pound, and the grafted monomer is a methacrylic monomer.

13. A composition as defined in Claim 8,
wherein said core shell polymer comprises units of
25 butadiene, styrene and a C₁-C₆ alkyl methacrylate.

14. A composition as defined in Claim 9,
wherein in said core shell polymer the C₁-C₆ methacrylate
is methyl methacrylate.
30

15. A composition as defined in Claim 1, wherein said aromatic polycarbonate resin includes units derived from bisphenol-A.

16. A composition as defined in Claim 1,
5 which also includes (d) a filler and/or reinforcing agent in an amount of from about 1 to about 60 parts by weight per 100 parts by weight of (a), (b) and (d) together.

17. A composition as defined in Claim 16,
10 in which (d) is glass, mica, talc or clay.

18. An improvement in the method of preparing a thermoplastic composition comprising admixing :

- (a) an aromatic polycarbonate resin; and
- (b) the balance to make 100 parts of (a) and (b) together
15 of an acrylic or methacrylic polymer of a conjugated diene, alone or in combination with a vinyl monomer, the core being polymerized from over 50 weight percent of said diene; and
- (c) a stabilizer composition comprising
 - 20 (i) a hindered phenolic antioxidant,
 - (ii) optionally, an aromatic amine,
 - (iii) a thioester, and optionally,
 - (iv) a trivalent phosphorus compound, said improvement comprising forming a preblend of components (b) and
 - 25 (c) and intimately admixing the preblend with component (a).

19. A thermoplastic composition made by the process of Claim 18.

20. A method as defined in Claim 1 wherein the
30 preblend of (b) and (c) is prepared by admixing the two in the presence of a small amount of a homopolymer or copolymer of ethylene.